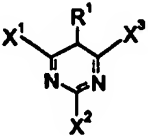




## INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

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<p>(54) Title: COMPOUND, COMPOSITION AND USE</p> <div style="text-align: center;">  <p>(1)</p> </div> <p>(57) Abstract</p> <p>There is described a negative charge control agent for use in electroreprography which comprises one or more pyrimidines of Formula (I), which includes all electroreprographically effective forms of such compounds selected from one or more of the following (including mixtures thereof and combination thereof in the same species): stereoisomers, zwitterions, polymorphs, solvates, isotopic forms; and salts thereof; where in Formula (I): R<sup>1</sup> represents a substituent selected from one or more of: H, hydroxy, mercapto, amino, nitro, sulphonyl, sulphino, carboxy, halo, cyano, optionally substituted C<sub>1-30</sub>hydrocarbyl, X<sup>1</sup>, X<sup>2</sup> and X<sup>3</sup> which may be the same or different, each independently represents a group capable of receiving a liable proton.</p>		

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COMPOUND, COMPOSITION AND USE

The present invention relates to the field of electroreprography. In particular the invention relates to compounds and compositions with utility in electroreprography (preferably as charge control agents); electroreprographic apparatuses comprising them; and methods for making these compounds, compositions and/or apparatuses.

5       Electroreprography is any process in which an image is reproduced by means of electricity and incident radiation, usually electromagnetic radiation more usually visible light. Electroreprography includes the technology of electrophotography which encompasses photocopying and laser printing technologies. In both these technologies a latent electrostatic image in charge is produced by exposure of a photoconductive drum to light. This can be either reflected light from an illuminated image (photocopying) or by  
10       scanning the drum with a laser usually under instruction from a computer (laser printing). Once a latent image has been produced in charge it must be developed with colorant so that a visible image can be printed onto paper.

      Toner compositions are used to develop the latent image on the drum into a visual  
15       image. During use in an electroreprographic device friction between particles of toner, their carrier and/or parts of the device in which the toner is used cause the toner particles to become charged with an electrostatic charge (tribocharge). The exact mechanism of development of the toner image will then vary according to the specific device used. For example in a conventional photocopier the toner composition may be formulated so that  
20       tribocharged toner particles will be opposite in sign to the latent image on the drum and toner will be attracted to the latent image on the drum to develop an image in toner on the drum which corresponds to the original document. The developed image is then transferred to a substrate such as paper (e.g. by a pressure roller and/or voltage). The transferred image is fixed to the substrate (e.g. by heat) to produce a hard copy of the  
25       image. The image drum is then cleaned and the device is ready to produce the next copy. Thus toner compositions are used both to develop the latent image on the drum and to produce the final hard copy.

      Thus it is desirable for toner compositions to comprise particles which can possess readily an electrostatic charge (tribocharge) so they can be attracted to the latent  
30       image on the drum to develop the latent image. Toners which readily tribocharge may also have the further advantage of facilitating rapid and more complete removal of any residual toner from the image drum (e.g. by electrostatic repulsion). This may improve image quality (by reducing ghost images from previous copies) and may reduce the cycle time between copies and thus increase the speed of copying.

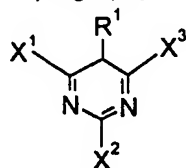
Toner compositions can exist in diverse physical forms such as dry toners which comprise particulate mixtures, or liquid toners in which the toner particles are dispersed within an insulating liquid. The toner particles of the required size can be formed by many different methods for example by physical methods such as grinding or milling particles ("conventional toners"), or by chemical production and growth of the particles ("chemically produced toners" - CPTs). Further examples of the requirements of typical toner compositions and examples thereof are given in the book "The Chemistry and Technology of Printing and Imaging Systems" edited by P. Gregory (published by Blackie Academic and Professional, 1996), especially in Chapter 4 entitled "Electrophotography".

It has been found that the addition of certain charge control agents (hereinafter known as CCAs) to toner compositions helps the production and stability of triboelectric charge within the toner. Use of CCAs may also lead to improved image quality when the latent image is transferred to the paper. The mechanism for the action of CCAs is unclear, but the industry continues to seek compounds with improved abilities as CCAs. Properties desired in ideal CCAs; toner compositions to which they are added; and/or the hard copies they produce are well known to those skilled in the art. Such properties might comprise any or all of the following: ability to stabilise larger tribocharge; improved tribocharge distribution and/or uniformity of charge within an individual toner particle and/or across the population of toner particles within a toner composition; reduced cost, reduced toxicity or non-toxicity, greater stability under conditions of use, good compatibility with the binder resin in a toner, improved image resolution, greater speed of image production, reduction in print bleed in the hard copy and/or improved colorant properties.

The CCAs currently available are not completely satisfactory in some or all of these respects. Thus it would be desirable to provide CCAs which result in improvements in some or all of the preceding areas.

The applicant has discovered that certain compounds may act as charge control agents and overcome some or all of the disadvantages with known CCAs. Preferred CCAs of the present invention are those which are substantially free of heavy metals, as these CCAs are easier to manufacture and may exhibit improved effects on the environment.

Therefore broadly in accordance with the present invention there is provided an electroreprographically effective form of at least one compound of Formula I as a charge control agent (CCA) for use in electroreprography,



Formula I

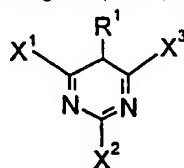
which includes all electroreprographically effective forms of such compounds selected from one or more of the following (including mixtures thereof and combinations thereof in the same species): stereoisomers, zwitterions, polymorphs, solvates, isotopic forms; and salts thereof;

the compound of Formula I being substantially free of impurities; and substantially free of heavy metals; and where in Formula I:

R¹ represents a substituent selected from one or more of: H, hydroxy, mercapto, amino, nitro, sulpho, sulpheno, sulphino, carboxy, halo, cyano, optionally substituted C<sub>1-30</sub>hydrocarbyl,

X¹, X² and X³ which may be the same or different, each independently represent a group capable of receiving a liable proton.

In a further aspect of the present invention there is provided a composition effective for use in electroreprography comprising particles of a toner, the composition further comprising as a charge control agent (CCA) at least one compound of Formula II:



Formula II

which includes all electroreprographically effective forms of such compounds selected from one or more of the following (including mixtures thereof and combinations thereof in the same species): stereoisomers, zwitterions, polymorphs, solvates, isotopic forms; and salts thereof; where in Formula II:

R¹ represents a substituent selected from one or more of: H, hydroxy, mercapto, amino, nitro, sulpho, sulpheno, sulphino, carboxy, halo, cyano, optionally substituted C<sub>1-30</sub>hydrocarbyl,

X¹, X² and X³ which may be the same or different, each independently represent a group capable of receiving a liable proton.

Preferably the compound of Formula II is substantially free of heavy metal, preferably any metal.

The term 'hydrocarbyl' as used herein denotes any radical moiety which comprises one or more hydrogen atoms with one or more carbon atoms and optionally one or more other suitable heteroatoms, preferably nitrogen, oxygen and/or sulphur. More preferably 'hydrocarbyl' moieties comprise any of the following moieties and combinations thereof in the same moiety: alkyl, alkoxy, alkanoyl, carboxy, alkanoyloxy, 5 alkoxy carbonyl, alkylthio, alkylsulphinyl, alkylsulphonyl, carbamoyl, sulphamoyl, alkylamino, and/or alkanoylamino. Hydrocarbyl moieties may also comprise one or more double and/or triple carbon to carbon bonds and/or aromatic moieties and may either in whole or in part be linear, branched and/or cyclic.

10 The term 'alkyl' or its equivalent (e.g. 'alk') as used herein may be readily replaced, where appropriate, by terms encompassing other hydrocarbon moieties such as those comprising double bonds, triple bonds, and/or aromatic moieties (e.g. alkenyl, alkynyl and/or aryl) as well as multivalent species attached to two or more substituents (such as alkylene). The term 'halo' as used herein signifies fluoro, chloro, bromo and 15 iodo, preferably fluoro and chloro.

Any radical group or moiety mentioned herein (e.g. as a substituent) refers to a monovalent radical unless otherwise stated or the context clearly indicates otherwise (e.g. an alkylene moiety is bivalent and links two other moieties). A group which comprises a chain of three or more atoms signifies a group in which the chain wholly or in part may 20 comprise one or more linear chains, branched chains and/or form rings (including spiro and/or fused rings). The total number of certain atoms is specified for certain substituents for example  $C_{1-n}$ hydrocarbyl, signifies an hydrocarbyl moiety comprising from 1 to n carbon atoms. In any of the formulae herein if one or more ring substituents are not indicated as attached to any particular atom on the ring [for example the substituent  $R^1$  in 25 Formula (1)] the substituent may replace any H attached to an atom in the ring and may be located at any available position on the ring which is chemically suitable.

Preferably any of the hydrocarbyl groups listed above comprise from 1 to 24 carbon atoms, more preferably from 1 to 18, most preferably from 1 to 10. It is particularly preferred that the number of carbon atoms in a hydrocarbyl group is from 4 to 30 10 inclusive.

The term 'optionally substituted' as used herein, unless immediately followed by a list of one or more substituent groups, means optionally substituted with one or more groups selected from: hydroxy, mercapto, amino, nitro, sulpho, sulpheno, sulphino, carboxy, halo and cyano.

35 Unless the context clearly indicates otherwise, as used herein plural forms of the terms herein are to be construed as including the singular form and vice versa.

The term 'heavy metal' as used herein denotes transitional metals and metals in the fifth period and above of the periodic table. Such metals may be environmentally and toxicologically undesirable.

5 The term "electroreprographically effective" (for example with reference to compounds of Formulae I and II) will be understood to comprise those substances which if used in electroreprography or any of the other uses specified herein provide the required properties to the composition and are compatible with the conventional carriers and/or diluents used to formulate electroreprographic compositions. For greater acceptance for use as a CCA compounds of Formulae I and II are preferably clear in the  
10 conventional Ames test for toxicology.

Preferred compounds of Formulae I and II described herein are those where R<sup>1</sup> is selected from H and C<sub>1-6</sub>alkyl; and X<sup>1</sup>, X<sup>2</sup> and X<sup>3</sup> are independently selected from amino (optionally N-substituted by one or more C<sub>1-30</sub>hydrocarbyl) and hydroxy.

15 More preferred compounds of Formulae I and II are those in which: R<sup>1</sup> is selected from H and C<sub>1-4</sub>alkyl; and X<sup>1</sup>, X<sup>2</sup> and X<sup>3</sup> are selected from hydroxy and amino.

Most preferred compounds of Formulae I and II are those in which: R<sup>1</sup> is H or methyl; X<sup>1</sup> is hydroxy, and X<sup>2</sup> and X<sup>3</sup> are different and selected from hydroxy and amino.

Specific compounds of Formulae I and II are selected from:

20 2-amino-4,6-dihydroxypyrimidine;  
4-amino-2,6-dihydroxypyrimidine;  
electroreprographically effective tautomer(s);  
electroreprographically effective salt(s); and  
electroreprographically effective mixture(s) thereof.

25 The substituents in compounds of Formulae I and II (e.g. R<sup>1</sup>) may be selected to improve the compatibility of the CCA with the resins with which they are formulated. Thus, the size and length of the substituents may be selected to optimise the physical entanglement or interlocation with the resin or they may contain reactive entities capable of chemically reacting with the resin.

30 Salts of Formulae I and II may be formed from one or more organic and/or inorganic acids and/or bases (for example acid and/or base addition salts). Salts of Formulae I and II comprise all acceptable salts that may be formed from monovalent and/or multivalent acids and/or bases (for example acid metal salts). Salts of Formulae I and II also comprise all enantiomeric salts formed with acceptable chiral acids and/or bases and/or any mixtures of enantiomers of such salts (for example racemic mixtures).

35 Preferred salts of Formulae I and II are those where the moiety of Formulae I and II comprises an anion. Preferred counter-ions in salts of compounds of Formulae I and II are cations, more preferably selected from one or more of: H<sup>+</sup> and ammonium cation

(optionally substituted with one or more hydrocarbyl groups which may be the same or different).

The counter-ion may also comprise one or more metal cation(s) such as suitable monovalent, divalent or trivalent metal cations. More preferably the metal cation(s) are selected from those groups 1a, 2a, 3a, 1b, 2b, 6b, 7b and 8 of the Periodic Table according to Mendeleef as, for example, published in the inside rear cover of the Handbook of Chemistry and Physics published by The Chemical Rubber Co., Ohio, USA. Most preferred metal cations are selected from one or more of Na, K, Mg, Ca, Zn and Al. However it will be appreciated that it is an advantage of a preferred embodiment of the invention is to provide compositions substantially free of heavy metals that may be associated with environmental and/or toxicological problems. Thus preferred compositions of the present invention are substantially free of any heavy metals, preferably all metals.

Salts of Formulae I and II may also be salts formed with a quaternary ammonium cation (hereinafter QAC). Preferred QAC's are those containing C<sub>1-30</sub> alkyl chains, particularly where the alkyl chain contains more than 6 and especially more than 10 carbon atoms since these QAC's are less volatile and are more resistant to the high temperature employed in the fabrication of toner resin compositions.

Suitable QAC cations may be selected from one or more of the following: N,N-diethyl-N-dodecyl-N-benzylammonium; N,N-dimethyl-N-octadecyl-N-(dimethylbenzyl) ammonium; N,N-dimethyl-N,N-didecyl ammonium; N,N-dimethyl-N,N-didodecyl ammonium; N,N,N-trimethyl-N-tetradecylammonium; N-benzyl-N,N-dimethyl-N-(C<sub>12-18</sub> alkyl)ammonium; N-(dichlorobenzyl)-N,N-dimethyl-N-dodecylammonium; N-hexadecyl pyridinium; N-hexadecyl-N,N,N-trimethylammonium, dodecylpyridinium; N-benzyl-N-dodecyl-N,N-bis(hydroxyethyl)ammonium; N-dodecyl-N-benzyl-N,N-dimethylammonium; N-benzyl-N,N-dimethyl-N-(C<sub>12-18</sub> alkyl)ammonium; N-dodecyl-N,N-dimethyl-N-(1-naphthylmethyl) ammonium and N-hexadecyl-N,N-dimethyl-N-benzylammonium cations.

Suitable QAC cations may also be formed from suitable amines for example from one or more amines selected from: dodecylamine, octadecylamine, didecylamine, didodecylamine, tetradecylamine, hexadecylamine, mixed C<sub>12-18</sub> alkylamines and N-benzyl amines. Preferred amines which may be used to form suitable QAC comprise N-C<sub>1-8</sub> alkyl primary amines, N,N-di-C<sub>1-8</sub> alkyl secondary amines and N-benzyl amines. Particularly preferred amines comprise methyl and ethyl amine derivatives.

Salts of Formulae I and II can include more than one counter-ion for example a combination of QACs as appropriate for the stoichiometry of a particular salt of Formulae I and II. It is preferred, however, that the counter-ion comprises only one cation.

Certain compounds of Formulae I and II may exist as one or more stereoisomers, for example enantiomers, diastereoisomers, geometric isomers, tautomers, conformers and/or combinations thereof if possible within the same moiety and/or residue (such as any suitable molecular and/or ionic species). In particular compounds of Formulae I and II may exist as many different tautomers. The present invention includes all electroreprographically effective stereoisomers (especially tautomers) of compounds of Formulae I and II and/or any mixtures thereof.

Certain compounds of Formulae I and II may exist as one or more zwitterions., for example where there exists two or more centres of ionic charge. The present invention includes all electroreprographically effective zwitterions of compounds of Formulae I and II and/or any mixtures thereof.

Certain compounds of Formulae I and II may exist as one or more polymorphs, for example different interstitial compounds, crystalline forms, amorphous forms, phases, solid solutions and/or any suitable mixtures thereof. The present invention includes all electroreprographically effective polymorphs of compounds of Formulae I and II and/or any mixtures thereof.

Certain compounds of Formulae I and II may exist in the form of one or more solvates formed from one or more electroreprographically acceptable solvents. The degree of solvation may be non-stoichiometric. Compounds of Formulae I and II may also exist in an unsolvated form (for example an anhydrous form). The present invention includes all electroreprographically effective solvated forms of compounds of Formulae I and II and/or any mixtures thereof.

Certain compounds of Formulae I and II may exist as one or more isotopic forms in which one or more of the commonly occurring isotopes of one or more atoms in compounds of Formulae I and II are replaced by an isotope of the same atom (for example a  $^{12}\text{C}$  atom may be replaced by a  $^{14}\text{C}$  atom). Optionally the isotopes may be radio-active. Certain isotopic forms of compounds of Formulae I and II may have utility as means for selective imaging in imaging devices (for example devices using X-rays, positron emission tomography and/or nuclear magnetic resonance); as tools to investigate the mode of action of compounds of Formulae I and II; and/or in any other uses suitable for isotopically labelled compounds of Formulae I and II. The present invention includes all electroreprographically effective, isotopic forms of compounds of Formulae I and II and/or any mixtures thereof.

The electroreprographic compositions of the present invention may further comprise a electroreprographically effective diluent and/or carrier (e.g. a suitable resin). Conveniently compositions of the present invention comprise toner compositions and developer compositions. Toner compositions may comprise particles of toner resin either as a powder composition (dry toners) or dispersed within an insulating liquid (liquid

toners). Developer compositions are similar to toner compositions but may further comprises large carrier particles (e.g. inert beads) which may support the smaller toner particles. Advantageously in the compositions of the present invention the compound(s) of Formulae I or II are present as the only charge control agent.

5       The present invention relates to compositions comprising any compounds of Formulae I and II even those which may not be directly electroreprographically effective. For example compounds of Formulae I and II which are ineffective for use in electroreprography (e.g. because they comprise a counter ion which is unsuitable) may have other utility such as an intermediate in the preparation and/or purification of  
10       electroreprographically effective compounds of Formulae I and II and/or as a research tool and/or diagnostic aid in relation to one or more of the uses described herein.

      The resin suitable for use as the diluent and/or carrier in compositions of the present invention (and/or which may form the toner resin particles) preferably comprises any thermoplastic resin suitable for use in the preparation of toner compositions. The  
15       terms resin and polymer are used herein interchangeably as there is no technical difference between them. More preferably suitable resins may be selected from one or more of the following: a styrene and/or substituted styrene polymer, (such as homopolymer [for example polystyrene] and/or copolymer [for example styrene-butadiene copolymer and/or styrene-acrylic copolymer {e.g. a styrene-butyl methacrylate  
20       copolymer}]); polyesters (such as specially alkoxylated bis-phenol based polyester resins [for example those described in US patent 5,143,809]), polyvinyl acetate, polyalkenes, poly(vinyl chloride), polyurethanes, polyamides, silicones, epoxy resins and phenolic resins. Further examples of these and other resins are given in the book "Electrophotography" by R. M. Shafert (Focal Press) and in the following patents or patent  
25       applications: GB 2,090,008, US 4,206,064, US 4,407,924 and US 5,230,926.

      Preferably compositions of the present invention comprise a CCA in amount from about 0.1% to about 12%, more preferably from about 0.5% to about 10% and most preferably from about 1% to about 3% by weight of the total composition.

      The compounds of Formulae I and II may be coloured or substantially colourless.  
30       Colourless CCAs have particular utility in coloured compositions which are not black (for example in toners having a light shade and/or colour) where colourless compounds would not substantially alter the colour of the composition to which they are added. If coloured preferably compounds of Formulae I and II are added to a composition in amounts insufficient to impart colour to that composition. As an alternative certain compounds of  
35       Formulae I and II in addition to acting as a CCA may also act as the colorant either alone or in combination with other colorants.

      The term 'colorant' as used herein encompasses both dyes (which are substantially soluble in the medium to which they are added) and pigments (which are

substantially insoluble in the medium to which they are added). A colorant comprises any material which imparts colour to a medium whether by scattering, absorption and/or reflection of some or all of electromagnetic radiation within the visible range.

Thus compositions of the present invention may comprise one or more suitable  
5 dyestuffs and/or pigments as colorant. Suitable colorants may be selected from one or more of carbon black, magnetite, metallised phthalocyanine, quinacridone, perylene, benzidine, nigrosine, aniline, quinoline, anthraquinone, azo disperse dye, benzodifuranone, metallised lake or pigment toner, water insoluble salts of a basic dye, and any mixtures thereof. The colorant may also be a water soluble basic dye, especially  
10 a triphenylmethane dyestuff. The composition may contain up to about 20% colorant and especially from about 3% to about 10% relative to the total weight of the composition.

When the colorant comprises magnetites and/or coloured pigment the colorant is preferably present from about 5% to about 70% and more preferably from about 10% to about 50% by weight of the composition. Mixtures of carbon black and magnetite are  
15 available commercially and those containing from about 1% to about 15% are preferred, especially those containing from about 2% to about 6% carbon black based on the weight of carbon black and magnetite.

Compositions of the present invention may be prepared by any method known to the art. One such method of preparing a toner composition typically comprises mixing  
20 suitable toner resin with one or more compounds of Formulae I and II and optionally other ingredients (e.g. those mentioned herein). Generally, this involves mixing the molten composition at temperatures from about 100°C to 200°C, in order to uniformly distribute the ingredients throughout the toner resin. The toner resin may then be cooled, crushed and micronised until the mean diameter of the particles is preferably below about 20µm  
25 and, for high resolution electroreprography, more preferably from about 1µm to about 10µm. The powdered colour toner or toner-resin so obtained may be used directly or may be diluted with an inert solid diluent such as fine silica by mixing for example in a suitable blending machine.

Compositions of the present invention may also comprise particles (e.g. of toner  
30 and/or developer) prepared chemically by agglomeration, coagulation and/or flocculation techniques. Chemically produced particles provide a greater degree of control of the properties of resultant particles such as size distribution, particle shape and/or particle composition.

Preferred compositions of the invention comprise toner particles having a mean  
35 size of below about 20 µm, preferably from about 3 to about 15 µm, most preferably from about 5 to about 10µm. The carrier, toner particles and/or diluent may comprise one or more resins as described herein.

Preferred compositions of the invention comprise a developer composition in which the carrier comprises larger particles having a mean size of between about 20µm and about 100µm.

5 The particle size given herein is a linear dimension corresponding to the diameter of a sphere approximately of same volume as the particular particle of interest which may be substantially irregular in shape.

Compounds of Formulae I and II may be produced by any suitable method, known in the art.

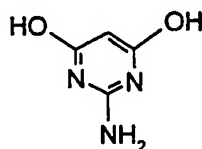
10 According to a further aspect of the invention there is provided use of compounds of Formulae I and II as a CCA. It is preferred that compounds of Formulae I and II act as CCAs which control negative electrostatic charge (hereinafter known as negative CCAs). More preferably negative CCAs of Formulae I and II are capable of being electrified with negative charge in use. Conveniently the compounds of Formulae I and II are neutral.

15 A still further aspect of the present invention provides an electroreprographic apparatus, component of the apparatus and/or consumable for use with the apparatus, which comprises one or more compounds of Formulae I and/or II and/or a composition as described herein.

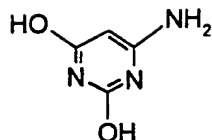
20 Another aspect of the invention provides use of one or more compounds of Formulae I and II and/or a composition as described herein; in the manufacture of an electroreprographic apparatus, component for the apparatus and/or consumable for use with the apparatus.

25 The invention is further illustrated by the following exemplified compounds which the applicant has found of use as CCAs in various toner and developer compositions. Each of the compounds below were prepared similarly to known methods. The crude products thus obtained were then purified to remove substantially all heavy metals and the compounds were used in their substantially pure form to prepare toners and developers as described below.

Example 1



30 2-Amino-4,6-dihydroxypyrimidine

Example 2

4-Amino-2,6-dihydroxypyrimidine

5 Preparation of toner compositions

The above compounds (in substantially pure form being substantially free of heavy metals) were used to prepare an electroreprographic pseudo toner according the formulations given below. The toner ingredients were mixed together until uniform. The mixture was then extruded at 160° C, ground into a coarse powder and milled using a jet mill to obtain toner particles. These toners are capable of being charged with a negative electrostatic charge.

Toner	CCA	Resin		Wax
		Type	Amount	
A	2	SA	96	2
A (comp)	0	SA	96	2

In the above table the amounts given are relevant parts by weight for each ingredient.

SA denotes a styrene acrylic resin and formulations "A (comp)" is a control formulation without any CCA.

15

Preparation of developers

An electroreprographic developing agent was then prepared from the above toners using the exemplified CCAs (as well as no CCA), by mixing the toner with a various carriers in a respective toner to carrier mass ratio of 2:98. The carriers were various conventional coated and uncoated iron and ferrite carriers available commercially from Powdertech. Corporation. They are denoted the table below by their Powdertech. reference codes. The tribocharge of these developers was determined using the standard blow-off method.

20

Results

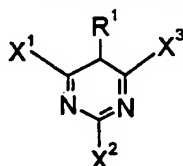
The results of tribocharge testing of the developers prepared as described above are given in the following table. The tribocharge values were all measured at the same time interval (30 minutes) after the initial charging of the developer:

CCA	Toner	Carrier	Tribocharge $\mu\text{C g}^{-1}$
Ex. 1	A	RAV 200	-20.4
Ex. 2	A	RAV 200	-44.3
None	A (comp)	RAV 200	-47.2
Ex 1	A	97-S7290	-16.8
None	A (comp)	97-S7290	-53.5

5 It can be seen that the exemplified CCAs stabilise the tribocharge of developers with a variety of different carriers compared to those developers without a CCA. After the initial charging, the tribocharge values of the developers comprising the exemplified CCAs were substantially time independent over the duration of the test. In comparison, developers without a CCA exhibit a lower (more negative) tribocharge value which increases throughout the test.

CLAIMS

1. An electroreprographically effective form of at least one compound of Formula I as a charge control agent (CCA) for use in electroreprography,



Formula I

which includes all electroreprographically effective forms of such compounds selected from one or more of the following (including mixtures thereof and combinations thereof in the same species): stereoisomers, zwitterions, polymorphs, solvates, isotopic forms; and salts thereof;

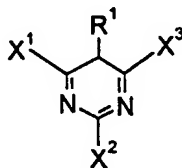
the compound being substantially free of impurities; and substantially free of heavy metals; and where in Formula I

R¹ represents a substituent selected from one or more of: H, hydroxy, mercapto, amino, nitro, sulfo, sulpheno, sulphino, carboxy, halo, cyano, optionally substituted C<sub>1-30</sub>hydrocarbyl,

X¹, X² and X³ which may be the same or different, each independently represent a group capable of receiving a liable proton.

2. A compound as claimed in claim 1, which is substantially colourless or weakly coloured.

3. A composition effective for use in electroreprography comprising particles of a toner, the composition further comprising as a charge control agent (CCA) at least one compound of Formula II



Formula II

which includes all electroreprographically effective forms of such compounds selected from one or more of the following (including mixtures thereof and combinations thereof in the same species): stereoisomers, zwitterions, polymorphs, solvates, isotopic forms; and all suitable salts thereof; where in Formula II:

R<sup>1</sup> represents a substituent selected from one or more of: H, hydroxy, mercapto, amino, nitro, sulpho, sulphenyl, sulphinyl, carboxy, halo, cyano, optionally substituted C<sub>1-30</sub>hydrocarbyl,

5 X<sup>1</sup>, X<sup>2</sup> and X<sup>3</sup> which may be the same or different, each independently represent a group capable of receiving a liable proton.

4. A composition as claimed in claim 3, in which in Formula II,  
R<sup>1</sup> is selected from H and C<sub>1-8</sub>alkyl; and  
X<sup>1</sup>, X<sup>2</sup> and X<sup>3</sup> are independently selected from amino (optionally N-substituted by one or  
10 more C<sub>1-30</sub>hydrocarbyl) and hydroxy.

5. A composition as claimed in either claim 3 or 4, in which in Formula II:  
R<sup>1</sup> is selected from H and C<sub>1-4</sub>alkyl; and  
X<sup>1</sup>, X<sup>2</sup> and X<sup>3</sup> are selected from hydroxy and amino.  
15

6. A composition as claimed in any of claims 3 to 5, in which in Formula II:  
R<sup>1</sup> is H or methyl;  
X<sup>1</sup> is hydroxy, and  
X<sup>2</sup> and X<sup>3</sup> are different and selected from hydroxy and amino.  
20

7. A composition as claimed in any of claims 3 to 6, in which the compound of  
Formula II comprises one or more of:  
2-amino-4,6-dihydroxypyrimidine;  
4-amino-2,6-dihydroxypyrimidine;  
25 electroreprographically effective tautomer(s);  
electroreprographically effective salt(s); and  
electroreprographically effective mixture(s) thereof.

8. A composition as claimed in any of claims 3 to 7, in which the toner particles have  
30 a mean size of below about 20µm.

9. A composition as claimed in any of claims 3 to 8, which further comprises carrier  
particles having a mean size of from about 20µm to about 100µm.

35 10. A composition as claimed in any of claims 3 to 9, which is substantially free of heavy metal.

11. Use as a charge control agent: of a substance comprising at least one of the following:

- a) a compound of Formula I as claimed in either claim 1 or 2 and/or
- b) a composition as claimed in any of claims 3 to 10.

5

12. A method for making a composition as claimed in any of claims 3 to 10, by mixing with a suitable carrier and/or diluent at least one compound of Formula I as claimed in either claim 1 or 2; and/or at least one compound of Formula II as represented in any of claims 3 to 10.

10

13. A method of manufacturing of an electroreprographic apparatus; and/or a component of the apparatus and/or consumable for use with the apparatus, the method using at least one of the following:

- a) a compound of Formula I as claimed in either claim 1 or 2; and/or
- b) a composition as claimed in any of claims 3 to 10.

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14. An electroreprographic apparatus, a component of the apparatus and/or a consumable for use with the apparatus, which comprises at least one of the following:

- a) a compound of Formula I as claimed in either claim 1 or 2; and/or
- b) a composition as claimed in any of claims 3 to 10.

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15. A compound, composition, electroreprographic apparatus and method for making any of the aforementioned, which is substantially as described herein with reference to the Examples.

## INTERNATIONAL SEARCH REPORT

International Application No

PCT/GB 98/03393

## A. CLASSIFICATION OF SUBJECT MATTER

IPC 6 G03G9/097 C07D239/54

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 6 G03G C07D

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	PATENT ABSTRACTS OF JAPAN vol. 9, no. 64 (P-343) '1787!, 23 March 1985 & JP 59 200253 A (FUJITSU), 13 November 1984 see abstract	1-5,8-15
X	PASTOR: "Synthesis and Structure of New Pyrido(2,3-d)pyrimidine Derivatives with Calcium Channel Antagonist Activity" TETRAHEDRON LETTERS, vol. 5027, 1994, pages 8085-8098, XP002092648	1,2
A	see page 8086; examples CPD,7,8	6,7
A	US 4 710 443 A (K.TANAKA) 1 December 1987 see claim 1	1-15



Further documents are listed in the continuation of box C.



Patent family members are listed in annex.

## \* Special categories of cited documents :

- \*A\* document defining the general state of the art which is not considered to be of particular relevance
- \*E\* earlier document but published on or after the international filing date
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- \*O\* document referring to an oral disclosure, use, exhibition or other means
- \*P\* document published prior to the international filing date but later than the priority date claimed

\*T\* later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

\*X\* document of particular relevance: the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

\*Y\* document of particular relevance: the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.

\*G\* document member of the same patent family

Date of the actual completion of the international search

8 February 1999

Date of mailing of the international search report

18/02/1999

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# INTERNATIONAL SEARCH REPORT

Inte: onal Application No

PCT/GB 98/03393

## C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	<p>PATENT ABSTRACTS OF JAPAN  vol. 17, no. 107 (P-1496), 4 March 1993  &amp; JP 04 296768 A (RICOH), 21 October 1992  see abstract</p> <p>-----</p>	1-15

# INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

PCT/GB 98/03393

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		JP 3056637 B	28-08-1991
		JP 61213855 A	22-09-1986
		JP 1701226 C	14-10-1992
		JP 3067266 B	22-10-1991
		JP 61258265 A	15-11-1986
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